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Variation of structure and magnetic properties with thickness of thin $Co_{59}Fe_{26}Ni_{15}$ films

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Abstract

Variations of phase composition and magnetic properties of electrodeposited nanocrystalline Co–Fe–Ni films with film thickness in the range of 50–500 nm were analyzed. The samples were magnetically soft with coercivity in the range $H_c = 2-20$ Oe and uniaxial magnetic anisotropy up to $H_k = 20$ Oe. It was found that H_c decreases and H_k increases with increasing film thickness. The BCC phase dominates at small film thickness up to about 80 nm and the FCC phase increases when the film growths to a larger thickness. The increase of FCC phase correlates with the improvement of the ultrasoft magnetic properties.

PACS: 75.75; 75.37.Nq

Keywords: Soft magnetic materials; Coercivity; Anisotropy; Thin films-thickness; Co–Fe–Ni; Nanocrystalline thin films; Microstructure; Phase composition

1. Introduction

So far, permalloy is commonly used in the production of thin-film magnetic read–write heads and of GMR read head shields [1,2]. However, Co–Fe-based alloys form promising alternatives because of a saturation magnetization M_s up to 2 T, i.e. much higher, than common permalloy layers. It was found that the most favorable soft magnetic properties occur in the materials with a dual phase of BCC and FCC. Among the factors defining the phase composition, the elemental relative concentration and deposition conditions are the most important [1,3–5].

We report here on the effect of thickness of CoFeNi films on microstructure, phase composition and magnetic properties. The results will be discussed in the context of the previously reported data.

2. Experimental

The CoFeNi layers were produced by electrodeposition at room temperature in the galvanostatic mode on copper underlayer (300 nm), which was sputtered on Cr-covered (20 nm) oxidized (200 nm) silicon [6]. The counter electrode was a plate of cobalt metal. The bath was mechanically stirred. A horizontal magnetic field of

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0.1 T, parallel to the plane, was applied in order to introduce in-plane uniaxial anisotropy. Different electrodeposition bath composition, current densities and temperature have been tested. The best results were obtained at room temperature with the current density of several mA/cm^2 in the bath: $CoSO_4 \cdot 7H_2O$—0.075 mol/l, $FeSO_4 \cdot 7H_2O$—0.020 mol/l, $NiSO_4 \cdot 6\,H_2O$—0.20 mol/l, H_3BO_3—0.40 mol/l, pH = 2.80–2.85, which was the same as in Ref. [5]. The magnetic coercivity, H_c, and anisotropy, H_k, were evaluated from hysteresis loops measured by VSM at room temperature with the magnetic field applied in the easy and hard directions, respectively, and varied in the range of ± 1 kOe [6].

The chemical composition of the layers was checked by means of SAM-EDAX, which resulted in $Co_{59}Fe_{26}Ni_{15}$ assignment. The structure of the deposited layer was determined by X-ray diffraction (XRD), using CuK_α radiation and transmission electron microscopy (TEM). From XRD and TEM we found that the layers consisted of mixtures of BCC and FCC nanocrystalline grains. The layers were strongly textured with dominant (1 1 0)BCC and (1 1 1)FCC XRD reflections at $2\vartheta = 45.25^\circ$ and $2\vartheta = 44.2^\circ$, respectively (see inset in Fig. 1). In all XRD measurements the samples were given an offset of 2° with respect to the bisector of the incoming and outgoing radiation in order to reduce reflections from the silicon single crystalline substrate. The XRD spectra were analyzed with the peak fitting program XFIT [7].

For TEM characterization the film surface first was covered by a layer of a glue, then the film was separated from the substrate by dissolution of Cu underlayer in a solvent, the film was fixed and finally the glue was removed by dipping it in acetone. The film was investigated using JEOL 2010F transmission electron microscope equipped with a postcolumn energy filter that provide an additional magnification of around 20 at the plane of the CCD camera with respect to the maximal attainable magnification by using a low-field objective minilens.

3. Results and discussion

The electrodeposited $Co_{59}Fe_{26}Ni_{15}$ films showed a nanocrystalline microstructure with a grain size from 10 to 30 nm as determined from the width of the (1 1 0)BCC and (1 1 1)FCC XRD peaks. The microstructure is shown in a bright-field TEM image in Fig. 1. The diffraction pattern in the inset to Fig. 1 consists of the rings, which also indicate a small grain size in a nanometer range. A high-resolution TEM image in Fig. 2 further illustrates the nanocrystalline microstructure of the film. The figure shows that the film is composed of grains with a size of 5–20 nm, separated by small- and large-angle boundaries, approximate positions of which are drawn as white lines. Some of the grains are highly disordered.

The samples were magnetically soft with coercivity in the range $H_c = 1-10$ Oe and uniaxial magnetic anisotropy up to $H_k = 20$ Oe [6]. The dependence of magnetic properties on the film thickness can be one of the reasons of variation of the H_c and H_k values from one sample to another. Considering a variation of the film thickness as a perturbation factor, Néel obtained his well-known "$\frac{-4}{3}$" dependence of the coercive field on the thickness,

$$H_c = Ct^n, \tag{1}$$

where $n \cong \frac{-4}{3}$ and $C = 0.71(\gamma_0^4/K^2 J_s)^{1/3}$ is a proportionality constant with γ_0 being the surface energy of the domain walls, K the anisotropy constant and J_s the saturation magnetization [8]. A thickness variation

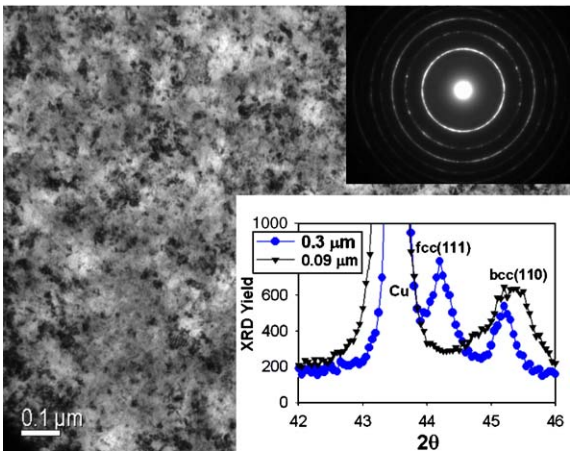

Fig. 1. In-plane TEM image of CoFeNi film. TEM and XRD diffraction patterns are shown in the inset.

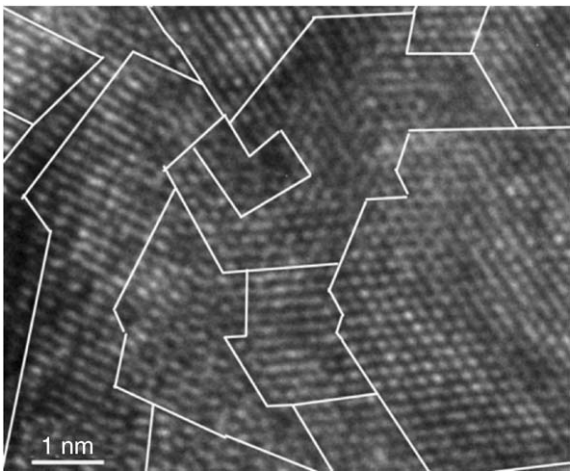

Fig. 2. HRTEM image showing the nanosized grains and defects. White lines were drawn by hand.

of H_c with somewhat different $n \cong -0.73$ was found in Ref. [5] for electrodeposited CoFeNi system with $t = 250-1000$ nm.

Our data, Fig. 3, extend those of Ref. [5] to smaller thicknesses and can be fitted by an expression

$$H_{ce} = H_{c0} + A/t \tag{2}$$

with $H_{c0} \cong 6.6$ Oe and $A \cong 0.6$ Oe/(mg/cm^2).

It was also noted by Néel that, although valid for the amplitudes of the thickness modulations, at least, comparable with the widths of domain walls, Eq. (1) does not contain the parameters of the modulations. Recently, the problem of coercivity dependence on the film thickness and roughness was considered by Ref. [9]. For the case when the film thickness is much larger than the domain wall thickness, expression (31) in Ref. [9] for the coercivity field of the Néel domain wall movement can be approximated by a very simple expression

$$H_c^{mov} \cong \pi(\pi A_{ex})^{1/2}(\rho_{rms}/t), \tag{3}$$

where A_{ex} is the exchange constant and ρ_{rms} is the root-mean-square of the local roughness slope. Assuming that the ρ_{rms} does not vary much in our range of thickness variation, we obtain from Eq. (3) the $1/t$ dependence which is in a qualitative correspondence with our experimental data for H_c, approximated by Eq. (2). The term H_{c0} in Eq. (2) that is due to pinning defects, preventing the domain motion, does not vary with increase of the thickness. Contrary to H_c, the anisotropy H_k tends to increase when the thickness increases (Fig. 3b).

As mentioned in Section 1, the variation of the phase composition influences the soft magnetic properties [1,2]. According to Ref. [4], the formation of a FCC + BCC mixture leads to the decrease of the grain size and, consequently, to the improvement of the soft magnetic properties. From Fig. 3a one can see that the BCC phase dominates at small film thickness of about 80 nm. However, the FCC fraction becomes as strong as the BCC phase at a thickness of about 200 nm. From Figs. 3a, b and 4 one can see that the increase of FCC phase correlates with the improvement of the ultrasoft magnetic properties, i.e. with the decrease of coercivity and the increase of uniaxial anisotropy. This observation is in line with those of Osaka for CoFeNi films. The main agent, which defines the mixture in the phase composition, is Fe: CoFeNi solutions with less Fe-content are FCC phase, and those rich with Fe have BCC phase. The change from FCC to FCC + BCC mixture to BCC was observed in electrodeposited FeNi alloy in Ref. [10] when the concentration of Fe increased, while for CoNi alloys, FCC, mixed FCC and HCP, and HCP as-deposited Co content increased. The smallest grain size was obtained in the mixed phase region of both NiFe and CoNi alloys.

The correlation between elemental concentration and coercivity was also observed in Ref. [5], both depending on the deposition condition (current density) and film thickness. The coercive field and Ni content decreased whereas the Co and Fe content increased when thickness increased from 0.25 to 1 μm. Since the higher concentration of Fe assumes the lower fraction of the FCC fraction in the alloy, one could expect a decrease of the FCC fraction with increasing thickness. Our data in Fig. 3a shows an opposite tendency of the FCC/BCC ratio as a function of thickness. Also, the decrease of H_c with increasing thickness in Ref. [5] could be reasonably connected with an increase of the FCC/BCC ratio, following the arguments of Ref. [4]. Finally, the observed depth variation of Fe in Ref. [5] is in contrast with observations with permalloy films deposition, where Fe content at the interface is usually higher than in the bulk [11]. The cause of this contradiction is not clear yet and might be connected with a more

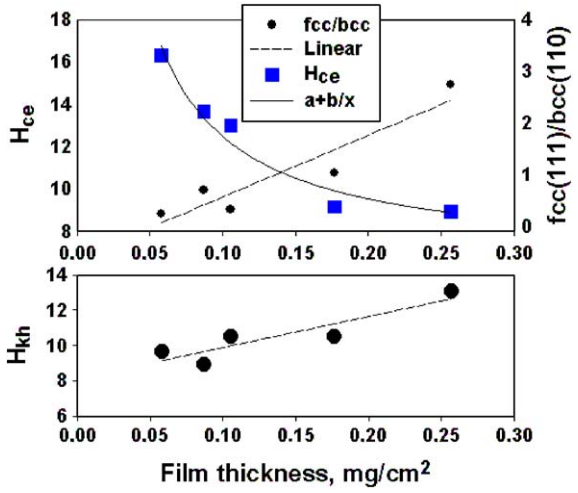

Fig. 3. Coercive (H_{ce}) and anisotropy (H_{kh}) fields and FCC/BCC ratio variations with the film thickness.

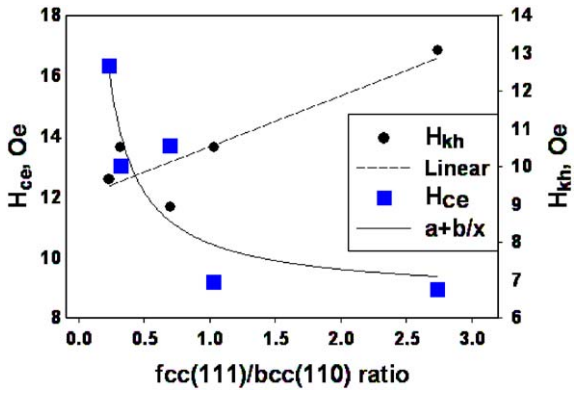

Fig. 4. Coercive and anisotropy field variation with the FCC/BCC ratio.

complicated influence of microstructural properties of the films.

4. Conclusions

We have extended the study of Ref. [5] on the thickness dependence of magnetic properties of electrodeposited Co–Fe–Ni films to a smaller thickness down to 50 nm. From the width of the main XRD reflections and TEM and HRTEM images the average grain size from 10 to 30 nm were estimated, depending on slight variations in the deposition conditions. We have shown that, in accordance with Refs. [5,8], the coercivity increases in the films with decreasing thickness. Quantitatively, the $H_c(t)$ dependence follows the treatment of Zhao et al. [9] where the surface roughness was taken into account. We have also shown that the FCC to BCC ratio varies with thickness of the film for our deposition condition. The BCC phase dominates at small film thickness of about 80 nm. However, the FCC fraction becomes as strong as the BCC phase at a thickness of about 200 nm. The uniaxial anisotropy has a tendency to increase with increasing film thickness.

Acknowledgments

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